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Ph.D. thesis 2005

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Preface

This thesis “Preferential Flow and Transport in Variably Saturated Fractured Media” has been submitted as a part of the requirements for the Ph.D. degree at the Technical University of Denmark (DTU). The work presented in this thesis was carried out at the Geological Survey of Denmark and Greenland (GEUS), Department of Quaternary Geology and Department of Hydrology respectively, in the period November 2000 to July 2005. A part of the study was done at Département de Géologie et de Génie Géologique, Université Laval, Québec, Canada. The work was partly funded by two EU-financed projects FRACFLOW (ENV4-CT97-0041) and TRACe-Fracture (EVK1-CT1999-00013), and the project “Concept for selection of Pesticide-sensitive areas” financed by the Danish Environmental Protection Agency.

The thesis includes four manuscripts, on which a synopsis summarize is written presenting four major subjects, where relevant literature including the Ph.D.-contribution is incorporated:

Manuscript I	Rosenbom A.E., and Jakobsen P.R. *IR Thermography and Fracture Analysis of Preferential Flow in Chalk.* Published in Vadose Zone Journal 4: 271-280, 2005.
Manuscript II	Rosenbom A.E., Ernstsen V., Flühler H., Jensen K.H., Refsgaard J.C., and Wydler H. *Fluorescence Imaging of Tracer Distributions in Variably Saturated Fractured Clayey Till.* Draft journal paper.
Manuscript III	Rosenbom A.E., Therrien R., Refsgaard J.C., Jensen K.H., Ernstsen V., and Klint K.E.S. *Numerical Analysis of Water and Solute Transport in Variably Saturated Fractured Clayey Till.* Draft journal paper.
Manuscript IV	Klint K.E.S., Gravesen P., Rosenbom A.E., Laroche C., Trenty L., Lethiez P., Sanchez F., Molinelli L. and Tsakiroglou C.D. *Multi-Scale characterization of fractured rocks used as a means for the realistic simulation of pollutant migration pathways in contaminated sites: A case study.* Published in Water, Air, and Soil Pollution: Focus 4: 201-214, 2004.

The papers are not included in this www-version of the thesis. The complete thesis can be obtained from the Library at the Institute of Environment & Resources, Bygningstorvet, Building 115, Technical University of Denmark, DK-2800 Kgs. Lyngby (library@er.dtu.dk) or the Geological Survey of Denmark and Greenland, Øster Voldgade 10, DK-1350 Copenhagen K (www.geus.dk).

Manuscript I is the result of a corporation with geologist and senior researcher Peter Roll Jakobsen, who taught me many things concerning fracture mapping in chalk and tectonic deformations. The work was performed within the framework of the EU-project FRACFLOW coordinated by the chalk-expert, geologist and senior advisor Erik Nygaard, whose guidance is very much appreciated. In addition, I would like to thank technicians Søren Nielsen and Erik V. Clausen for their assistance with the experimental work. Finally I would like to gratefully acknowledge senior-translator Marianne Frediani for commenting on the manuscript before submitting it.

Manuscript II is the result of a corporation with Professor Hannes Flühler and technician Hannes Wydler from ETH Zürich, Institute of Terrestrial Ecology (ITÖ), Soil Physics, Schlieren, Switzerland. After visiting ETH twice and being introduced to a special developed imaging device from ETH, Professor Hannes Flühler generous accepted collaboration on "Flourescence Imaging of Tracer Distribution in a Danish fractured clayey till". The fieldwork was performed within the project "*Concept for selection of Pesticide-sensitive areas*" where project-colleagues Dr. Vibeke Ernstsen, Dr. Ole Hørbye Jacobsen, Dr. Bo Vangsø Iversen, Dr. Knud Erik Klint and Dr. Søren Torp detailed description of the fractured till contributed to the overall understanding of the flow and transport pattern in the till. The fieldwork would not have been possible without the help of the three technicians Henning Hougaard, Per Jensen and Hans Jørgen Lorentzen. Especially Per was always there to assist during the long field days. Finally I would like to gratefully acknowledge agriculturalist and electrical engineer Joachim Noergaard for the design and the manufacturing of the sprinkler-system and for his guides and help in the field.

Manuscript III presents a numerical understanding of the results given in manuscript II. This work is the result of a corporation with Professor Rene Therrien from Département de Géologie et de Génie Géologique, Université Laval, Québec, Canada, who developed the model FRAC3DVS (now HydroSphere), which was used. Rene's visit to Denmark was partly funded by The Danish Oil and Gas Company, DONG. As a part of the Ph.D. requirements, I spent 2 months at Département de Géologie et de Génie Géologique, Université Laval, Québec, Canada, working with HydroSphere and setting up a temporary, not complete model based on my fieldwork data. I would like to thank Rene, his family and his research group for making my stay there very pleasant.

Manuscript IV presents work performed within the framework of the EU-project TRACe-Fracture. I contributed in the planning of the project, was responsible for planning the fieldwork, collecting the fracture samples in the field and prepare samples for mechanical fracture aperture measurement in the Scanning Electron Microscope (SEM) in the laboratory and also carried out the SEM-analyses of the samples and described the micromorphology of the fractures and faults. This work was performed in close corporation with Dr. Knud Erik S. Klint, senior advisor Niels Springer, student Morten Hansen and technician Hans Jørgen Lorentzen. At the Spanish field site, I especially

appreciated Hans Jørgen's assistance and knowledge concerning epoxy. Morten Hansen was a valuable help and guide with the Scanning Electron Microscope (SEM).

I gratefully acknowledge my three advisors: senior scientist Dr. Vibeke Ernstsen, Professor Karsten Høgh Jensen and Professor Jens Christian Refsgaard for their competent guidance and unfailing support. Special thanks are directed to Dr. Johnny Fredericia and Dr. Knud Erik S. Klint whose work within the fracture research inspired and formed the base for this Ph.D.-study. Also thanks to the rest of my colleagues at GEUS for creating a very nice atmosphere. Temporary head advisor at the Technical University of Denmark (DTU) Dr. Torben Sonnenborg and later Dr. Flemming Larsen are thanked for their support/help in the final stage of my PhD-study. Also Head of secretariat Anne Harsting at Environment & Resources, DTU, is acknowledged for her guides in the DTU-Ph.D.-study-administrative world.

Most of all, I wish to thank my family for their patience and support throughout the study-period, in which my son Ulrik and daughter Salla were born.

GEUS, July 2005, Annette Elisabeth Rosenbom

Abstract

A significant number of subsurface environmental problems involve preferential flow and solute transport in variably saturated fractured media. The presence of large connected void spaces such as fractures and channel-like openings in the vadose zone have been widely documented to provide rapid pathways for spreading of contaminant from the surface to deeper aquifers.

The objective of this Ph.D.-study is to characterise and estimate preferential flow and solute transport in three different variably saturated fractured media: chalk, till, and granite. All three media consist of relatively low permeable matrix with fractures and macropores introducing fast preferential flow and transport paths. In these media, not all the fractures and macropores are registered to be of equal importance for the flow and transport regime. The most important hydraulic pathways and hereby the preferred pathways are thus identified through detailed fracture mapping, an identification of the sources or mechanisms that are capable of producing the stress field requisite for initiating and propagating the fractures, and detailed in situ tracer-experiments.

The standard procedure for monitoring leaching processes is based on indirect methods such as pumping tests, well interference studies and tracer tests, which include extraction of soil solutions using suction cells or extraction of soil samples in situ. The limitations by using these procedures are that they only provide aggregated information on the effects of the fractures and macropores at scales significantly larger than the fractures and macropores because of the data uncertainty on fracture and macropore locations, properties, connectivity, boundary conditions, fracture- and macropore-matrix interactions, and host matrix properties. To circumvent these limitations, detailed tracer tests combined with detailed characterisation of the fractured vadose chalk and till were performed.

For the chalk, infrared thermography (IRT) provided the refined spatial information to clarify the hydraulic activity of the five fracture-systems registered by analysis of the regional setting and fracture mapping along quarry walls and in wells. By making use of the contrast between the constant temperature of the deeper groundwater and the temperature of the exposed wall in the quarry on a cold winter and a hot summer day, zones of groundwater discharge were delineated.

For the till, tracer experiments with two fluorescent tracers Acid Yellow 7 (AY7) and Sulforhodamine B (SB) were performed with three different rain events in a fall and summer season. The movement of both tracers in exposed profiles was delineated simultaneously by high spatial resolution concentration maps obtained with an imaging device. Additionally, a detailed description of the fractured vadose media including fracture and macropore mapping and estimation of structural properties of the different domains (biopores, fractures, coating, oxidized and reduced matrix) was performed. The

two-dimensional concentration distribution profiles of the tracers showed that: (a) biopores dominated the tracer migration in the upper 1.2 meter, (b) dead-end biopores were not activated in the fall season, (c) only tectonic fractures connected to hydraulically active biopores contributed to the migration, and (d) the water content in the upper 20 cm of the irrigated till had a pronounced retardation effect on the migration of AY7 but no effect on the migration of SB.

To perform risk assessment analysis on these naturally fractured variably saturated media, a better understanding of the often-controlling effects of the media is vital. A conceptual model based on detailed field-scale information concerning the different domains and a numerical model tool capable of representing the processes taking place in the media are in this connection crucial.

For the till, the three-dimensional numerical model, HydroSphere, describing fully integrated subsurface and solute transport was used to interpret the fluorescent tracer experiments. Special emphasis was given to address the different tracer migration pathways given the initial water content, rain intensities, tracer characteristics, geometry of the structure system, and domain properties.

For the granite, a description of the micromorphology of the fractures and faults and measurement of the fracture aperture (scale~0.01-1 mm) was conducted based on detailed analysis of impregnated fractured granite-samples in a Scanning Electron Microscope (SEM). Others used this information as input to the three-dimensional numerical model, SIMUSCOPP, for the simulation of a transient NAPL migration scenario based on a multi scale geological investigation (multi-scale fracture properties).

Resumé (Summary in Danish)

Et signifikant antal miljø-problemer involverer præferentiel strømning og stof transport i opsprækkede umættede medier. Tilstedeværelsen af store forbundne hulrum såsom sprækker og kanal-lignende åbninger i den umættede zone har længe været kendt for at tilvejebringe hurtige veje for spredning af forureninger fra overfladen til grundvands-magasiner.

Formålet med dette Ph.D.-studium er at karakterisere og estimere de præferentielle strømnings- og transport-veje i tre forskellige opsprækkede medier: kridt, moræneler og granit. Alle tre medier er karakteriseret ved relativ lavpermeable matrix med sprækker og makroporer, som introducerer hurtige præferentielle strømnings- og transport-veje. I disse medier er alle sprækkerne og makroporerne af samme vigtighed for strømning og transport regimet. De vigtigste hydrauliske veje er identificeret igennem detaljerede opmålinger af sprækker, identifikation af kilder og mekanismer, som er i stand til at udvikle et stres-felt nødvendig for at initiere og forplante sprækker, samt detaljerede in situ tracer-forsøg.

Standard proceduren til brug ved monitering af udvasknings-processerne er baseret på indirekte metoder såsom pumpetests, borings-interference studier og tracer tests, der inkluderer ekstraktion af jordvæsker ved brug af sugeceller eller ekstraktion af jordprøver in situ. Begrænsningerne ved brug af disse metoder er, at de kun tilvejebringer aggregeret information vedrørende effekten af sprækker og makroporer på en skala signifikant større end sprækkerne/makroporerne. Dette skyldes datausikkerhed på lokalisering, egenskaber, forbindelse, randbetingelser af sprække og makroporer, sprække- og makropore-matrix interaktion og matrix egenskaber. For at omgå disse begrænsninger er detaljeret tracer-tests blevet kombineret med detaljeret karakterisering af sprækket umættet kridt og moræneler.

For kridtet har infrarød termografi (IRT) tilvejebragt raffineret rumlig information til afklaring af den hydrauliske aktivitet af de fem sprække-systemer registreret ved en analyse af den regionale opsætning samt sprækkeopmåling langs kridtbruddets vægge og i boringer. Ved at udnytte kontrasten mellem den konstante temperatur af dybere grundvand og temperaturen af den blotlagte væg i bruddet på en kold vinter og en varm sommer dag er zoner med grundvandsudstrømning skitseret.

På moræneler er der foretaget tracer eksperimeter med to fluorescerende tracere Acid Yellow 7 (AY7) og Sulforhodamine B (SB) ved tre forskellige regnhændelser på to årstider (efterår og sommer). Udbredelsen af tracere er skitseret simultant i blotlagte profiler ved koncentrationskort af en høj opløselighed indhentet ved brug af et specielt designet foto-udstyr. Yderligere er der foretaget en detaljeret beskrivelse af det opsprækkede umættede medie inkluderende sprækker/makroporer kortlægning og estimering af strukturelle egenskaber for de forskellige domæner (bioporer, sprækker, belægning,

oxideret og reduceret matrix). De todimensionale koncentrations-fordelings-profiler af tracerne viser, at: (a) bioporerne dominerer tracermigrationen i de øverste 1.2 meter, (b) ”dead-end” bioporer ikke var aktive om efteråret, (c) kun sprækker forbundne med hydraulisk aktive bioporer bidrager til migrationen og (d) vandindholdet i de øverste 20 cm af den overrislet moræneler har en markant forsinkende effekt på udbredelsen af AY7, men uden effekt på udbredelsen af SB.

For at kunne foretage risiko-analyse på disse naturlige opsprækkede variabelt mættede medier, er en bedre forståelse af de ofte kontrollerende effekter af mediet vital. En konceptuel model baseret på detaljeret feltskala informationer omhandlende de forskellige domæner og en numerisk model i stand til at repræsentere processerne, som finder sted i mediet, er i denne sammenhæng af afgørende betydning.

For morænelers-aflejringen blev den tredimensionale numeriske model HydroSphere anvendt til at fortolke udbredelsen af fluorescerende tracer. Særlig vægt er der lagt på at kunne adressere de to tracers forskellige migrationsveje ved det initielle vandindhold, regn intensiteter, tracer karakteristik, geometrien af struktur-systemet samt domæne egenskaber.

For granitten, blev mikromorfologien af sprækker og forkastninger beskrevet samt deres apertur målt (skala~0.01-1mm) ved detaljeret analyse af imprægnerede opsprækkede granit-prøver i et Scan Elektron Mikroskop (SEM). Andre anvendt efterfølgende denne information som inddata til en tredimensional numerisk model, SIMUSCOPP, til at simulere et transient NAPL migrations scenarium baseret på en multi-skala geologisk undersøgelse (multi-skala sprække egenskaber).

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Introduction

Motivation

The problems with contaminated areas were launched with the development of the modern industrial and consumption-oriented society and the formerly sometimes careless use of chemicals and means of production in industry and commerce. In the late 70s, the general public is becoming more aware of the problems with contaminated areas. An increased environmental awareness, new findings on the behaviour of pollutants in the environment and developments in measuring techniques are additional factors, which have contributed to the realisation of the problems related to contaminated areas during the past few years. The relationship between the soil and important spheres of human interest such as public health, food and drinking water supply and other environmental issues is becoming increasingly apparent. More and more countries have now realised that a national policy is necessary to deal with the variety and complexity of problems associated with soil contamination.

Many of these environmental problems involve fluid flow and solute transport in the vadose zone. A zone, which refers to the part of the subsurface from land surface down to the lowest seasonal water table elevation. The zone may be composed of lithologies such as consolidated rock and/or unconsolidated granular material. Sites that are situated on lithologies with medium to low bulk permeability and very low matrix permeability pose a special problem. These lithologies are typical fractured rocks, tills and chalks, which covers a major part of the European countries. It has been widely documented that fractures and macropores, such as burrows and root-holes, provide rapid pathways for transport of contamination from contamination sources to the water table. Moreover fractures result in contaminant transport in unexpected directions depending on the fracture planes that are intersected, a very complicating factor in relation to predicting the fate and to the remediation of the contamination. The ability to reliably predict the rate and direction of preferential flow and contaminant transport in fractured variably saturated media and hereby to understand the processes of these systems would be of great economic benefit for planning and implementation of the remediation of contaminated fractured sites.

Objective

The objective of this Ph.D.-thesis has been to investigate preferential flow and transport in variably saturated fractured till, chalk and granite by a combination of detailed in-situ characterisation of the media and numerical modelling.

Major Subjects

Preferential Flow Pattern in Variably Saturated Fractured Media

Background

In lithologies flow can either be uniform (Green and Ampt, 1911) or non-uniform (Lawes et al., 1881). The uniform flow leads to a stable wetting front parallel with the soil surface and the non-uniform flow (un-even) results in irregular flow pattern, water moves faster and with increased quantity at certain locations in the vadose zone than at others. This non-uniform movement of water and dissolved solutes is commonly denoted preferential flow (Beven, 1991) and is characterised by its non-equilibrium nature (Simunek et al., 2003). This non-equilibrium feature is defined as a flow regime in which "for various reasons, infiltrating water does not have sufficient time to equilibrate with slowly moving resident water in the bulk of the soil matrix".

Preferential flow in natural media is generally recognized to arise from three factores: a) the presence of large connected void spaces, such as fractures, fissures, cracks, and macropores (decayed root channels and earthworm burrows) (Beven and Germann, 1982; Jørgensen et al., 1992; McKay et al., 1997; Villholth et al., 1998), b) the development of flow instability ("fingering") caused by profile heterogeneities, water repellence and/or air entrapment (Hendrickx et al.,1993; Glass and Nicholl, 1996), and c) "funnelling" of flow due to the presence of sloping layers that redirect the downward water movement (Kung, 1993).

In the subsurface, processes such as shrink-swell, freeze-thaw, biological activity (leading to development of biofilm (Vayenas et al., 2002), earthworms burrow and root channels), and physical manipulation (e.g., ploughing of an agricultural field) can dynamically alter the preferential flow pattern (National Research Council, 2001). These processes and their dynamics tend to decrease with depth, leaving the tectonic features/fractures to predominate (Klint and Gravesen, 1999). Additionally, all fractures are not of equal importance for the flow regime, and they may furthermore exhibit a dynamic hydraulic activity (Commission on Geoscience, Environment and Resources, 1996).

In recent years, several studies have produced evidence that vadose-zone flow in fractured media occurs at higher velocities than simple capillary-based models would predict. Pruess (1999) defines the fundamental paradox of unsaturated fracture flow: how can fast flow occur in the presence of the strong matrix imbibitions of variably saturated media?

Instability and the resultant gravity "fingering" is observed in unsaturated fractures, even when the matrix is unsaturated. In a tube (e.g. earthworm burrow) gravity fingering is expected to occur above a diameter defined by capillary properties of the system and at flux rate less than saturated flux (Glass and Nicholl, 1996). The "fingering" changes rapidly over time and undergoes cycles of snapping and reforming denoted intermittent flow, which is not considered by classical theory. These instable small-scale flow processes can substantially impact the transport in these features (Su et al., 2001).

Recently, the concept of film flow was introduced as a possible process by which preferential flow could occur on unsaturated fracture surfaces. "Films" as defined by Tokunaga et al. (2000) are a complex network of thick pendular regions that form within topographic depressions and thin films on topographic ridges. This transient film flow on rough fracture surface is capable of sustaining fast flow (average velocities greater than 10 my^{-1}) for average film thicknesses greater than 2 μm and matrix potentials greater than -10 cm H_2O. They also showed that the average film thickness dependence on matrix potential is approximately as a power function ($f=1.13\cdot(-h_m)^{-0.370}$, f = film thickness [μm] and h_m = matrix head [m]) and that the film hydraulic diffusivity increases with increased film thickness and matrix potential. Dragila and Wheatcraft (National Research Council, 2001) suggest that free-surface film flow is likely to occur in fractures larger than approximately 1 mm, because less energy is required to transport fluid that contacts one fracture wall compared with the transport of fluid that contacts both fracture walls. Free-surface film flow, which is only under the forces of gravity (including adsorptive surface forces) and not affected by capillary forces, can also occur in small-aperture as well as large-aperture fractures, flow rate is not directly controlled by the width of the fracture aperture, and the insert of flow does not require a contiguous fluid-filled pathway within the fracture. If the permeability of the fracture-coating is significantly greater than that of the matrix, fast "surface-zone flow" can occur even when the fractures are at very low water saturation (Tokunaga and Wan, 2001). Phillips et al. (1989), Tofteng et al. (2002) and Gjettermann et al. (2004) observed that flow through artificial macropores could occur as a water film along the macropore walls (film flow) or as moving water segments separated by air bubbles (pulse flow) even under unsaturated conditions. Ghodrati et al. (1999) showed that macropore flow occurred predominantly at the matrix-macro-pore interface and in the matrix zone directly adjacent to the macropore. Along this interface, only film flow was observed.

Fracture-matrix interaction plays a central role in the unsaturated flow pattern in the fractured porous media. Strong fracture-matrix interaction factors include: high matrix suction, large contact area between water and fracture wall, and absence of fracture coatings that impede matrix imbibition. Under these conditions, water in the fracture will be quickly absorbed into the matrix, and fracture flow cannot be sustained. By contrast, factors that create “weak” fracture-matrix interactions include: low matrix suction, small contact area between water and fracture wall, and presence of fracture coatings that impede matrix imbibition, Figure 1 (Thoma et al., 1992, Rosenbom et al., 2001). Under these conditions, water in the fracture is not readily absorbed into the matrix, and fracture flow may occur to significant depths (National Research Council, 2001). A similar interaction can be described for the macropore-matrix interaction. A number of investigators have reported that dry soil conditions tend to increase the contribution of macropores in infiltration and percolation and they have suggested that this was due to hydrophobicity of surface litter and soil and the openings of cracks (Sihipitalo et al., 1996). Macropores generated by the earthworm species *Lumbrius terrestric* (1-10 mm in diameter) are for example lined with mucus rich secretions, which is likely to make the burrow surface only partly hydrophilic (Lee, 1985). Shipitalo et al. (2004) have measured infiltration rates in individual *Lumbrius terrestric* burrows to range from 6 to 1043 ml min^{-1} in a clayey subsurface-drained soil. Contra dictionary, Flury et al. (1994, 1995) noted, that in well-structured soils there was a tendency for water to move deeper under wet than under dry conditions.

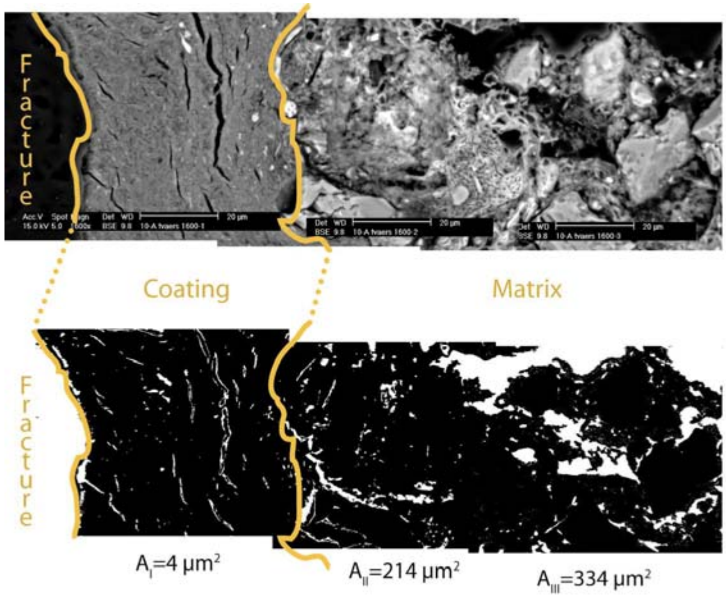

Figure 1. SEM- and binary-images (1600x) of a fracture zone with coating in a till-unit (Rosenbom, 2001).

Contribution

In manuscripts I, II and III, preferential flow in variably saturated fractured chalk and till is examined. In both cases, the presence of large connected void spaces was a controlling factor of the flow pattern in these settings.

In the relatively impermeable chalk it was possible to map the hydraulically active fracture systems based on a combination of infrared termography, detailed fracture mapping and existing regional tectonic and hydrologic knowledge. To obtain information on fracture settings at two different scales through the combined use of mapping techniques at field scale and analysis of regional setting has to my knowledge not previously been reported in the literature. By using this method, a clear link between the regional fracture setting and on site hydraulic active fracture systems was found emphasising the need for identification of the origin of the fractures.

A more complex preferential flow setting was found for the till. The porous variably saturated matrix was interacting to some degree with coated biopores and fractures. By irrigating a till-unit, water entered the variably saturated till-matrix but seemed to drain quickly into the biopores (primarily generated by earthworm). The water was then build up in "dead-end" biopores seeping into the matrix at relatively dry initial conditions (summer season) or migrating further into connected fractures to at least a depth of 3 meters below surface both at initially dry and wet conditions (fall and summer season) except for the 1 year rain event under wet initial conditions. Under this event, less water seemed to penetrate the macropores and matrix system. These results demonstrate even for high matrix suction (ψ<-100 cm H_2O), that biopores are activated hydraulically and that a fast flow mechanism in the biopores is present. It indicates hydrophobicity of surface litter and soil (Sihipitalo et al., 1996). These finding disagree with previously used concepts for macropore flow (Jarvis et al., 2002; Tokunaga et al., 2000). Also the fact, that only tectonic fractures connected with biopores seem to control the deep penetration, has not been visualised before. To neglect the fast flow mechanisms in macropores under high matrix suction and not to address the connection between biopores and tectonic fractures can therefore significantly underestimate the downward migration of contaminants in clayey till aquitards into underlying drinking water aquifers for planning, regulatory, and remediation purpose.

Solute Transport in Variably Saturated Fractured Media

Background

The flow regimes ability to transport solutes (e.g., natural constituents, artificial tracers, or contaminants) in fractured media is influenced by physical processes (advection, mechanical dispersion, diffusion), chemical processes (reactions between the solute and the solid material of the matrix and the fracture walls including sorption), and microbial processes (will not be described further).

Advective transport (advection) is the process by which solutes are transported along a path at an average rate equal to the average linear velocity of the water equal to the hydraulic conductivity divided by the effective porosity multiplied with the head gradient. Spatial spreading of solutes from this path is described as hydrodynamic dispersion. It occurs because of mechanical mixing during fluid advection (mechanical dispersion comprised of longitudinal and transverse dispersion) and because of molecular diffusion due to the thermal-kinetic energy of solute particles (Brownian motion).

The mechanical dispersion account for: 1) microscale spreading because of the parabolic velocity distribution in single pores, 2) variability in velocities between different pores, and 3) the tortuousity, branching and interfingering of pore channels (Freeze and Cherry, 1979). Here the longitudinal dispersion is a result of differences in travel time along flowlines, which split at grain boundaries (or large obstacles), whereas transversal dispersion applies to dispersion perpendicular to that direction and is caused by variations in the microscopic velocity within each flow channel and from one channel to another. Vertical transverse dispersion is usually smaller than horizontal transverse dispersion. Microscopically there is no mixing; however, if the average concentration of a given volume of fluid is considered an apparent dilution or spreading is present (Otaga, 1970). The mechanical dispersion in a variable aperture fracture is caused by Taylor dispersion, which results from velocity variations across the fracture aperture, and macro/geometric dispersion, which is caused by velocity variations in the planar of the fracture because of aperture variability (Detwiler et al., 2000). Since Taylor dispersion is proportional to v^2 and macro dispersion is linear proportional to v, where v is the mean flux $[LT^{-1}]$, mechanical dispersion will be negligible at Peclet numbers << 1. Dispersion takes effect at many scales, from pore-scale to larger scale. Variability in groundwater velocities may increase at larger scales for two reasons – either new, infrequently spaced, pore elements with higher-than-average velocities may be countered as scale is increased, as in fractured media, or there may be continuous variations in ensemble means from place to place, as in cases where Darcian permeability is inhomogeneous.

Molecular diffusion within the fracture is usually considered to be unimportant relative to mechanical dispersion. For a matrix with stagnant water, however, diffusion is the main process by which solutes are assumed to move. Matrix diffusion is believed to be an intrinsic mechanism for the exchange of mass between the matrix and the fracture (Maloszewski and Zuber, 1993; Jardine et al., 1999). It is controlled by (1) the local concentration gradient, (2) the molecular diffusion coefficient of the solute, and (3) the porosity and tortuosity of the matrix. Different formulations and solution methods for matrix diffusion have been suggested, all are though based on Fick's law (e.g. Tang et al., 1981). Diffusion in and out of the matrix will be significantly affected by the nature of the surface of the fracture. A thin layer of low permeability (coating/"fracture skin") can impede the interchange of water and solutes between the fracture and the matrix. In addition to the matrix, regions of immobile fluid may exist within the individual fractures (if flow through the fracture is channelled) and in "dead-end" fractures for a network. The effects of diffusive mass transfer or matrix diffusion on solute movement in laboratory structured porous media are well documented in the literature (Gwo et al., 2005).

Chemical processes of the geological setting may include equilibrium or time dependent sorption (Brusseau, 1994), cation exchange and anion exclusion (James and Rubin, 1986; Gvirtzman and Gorelick, 1991; Porro and Wierenga, 1993). In many cases these chemical processes are of much greater importance than hydraulic processes for the distribution of contaminates (Thomasson and Wierenga, 2003). These processes together with pH and redox potential of the soils and sediments are important properties for the fate of contaminants (Bodin et al., 2003). Together with diffusion, sorption plays a role in the retardation and retention of solutes in geological settings. Consequently, it is important to distinguish between diffusion and sorption. Since matrix diffusion is a relatively slow mechanism, sorption processes are mainly dominating at the fracture surfaces when flow is fast. Conversely, for significant mean resident times, sorption reactions mainly occur within the matrix because the available exchange surfaces in the matrix are much more important than those in the fracture. The phenomenon anion exclusion, which can be described by enhanced dispersion, effects a larger average transport velocity of dissolved anions through media than that of the accompanying water molecules because of electrostatic repulsion by negatively charged solid surfaces, which forces the anions away from the pore walls to areas with higher velocity.

Contribution

In manuscript II, the solute transport pattern of two fluorescent tracers AY7 and SB in fractured vadose till were delineated in details for three different rain events and two different initial water content distributions. The field experiment demonstrated that: (1) AY7 and SB were primarily transported in the biopores in the upper 1.2 meters, (2) the tracers were not detected in “dead-end” biopores when the matrix potential was near-zero – here only biopores connected to fractures were active in the tracer transport, and (3) piston-like migration of AY7 was observed for high matrix suctions while SB only migrated into the biopores. The results also demonstrated that matrix-fracture and -macropore interaction was negligible except in the upper 20-30 cm of the irrigated till-unit. Detailed delineation of transport paths in the upper 3 meters of a variably saturated fractured clayey till has so far not been reported from other field sites.

The reason for the difference in migration pattern of the two tracers is most likely related to differences in the surface charges of the tracer molecules. SB has a negative and a positive charged group, while AY7 has only a negative charged group. For the geochemical zone in question, free iron oxides with a positive charge, are present. Hence, the SB transport may be affected by cation exclusion from the iron oxides (and till matrix) because of its positively charged group and consequently SB is prone to preferential transport in the biopores. These pathways have a coating of clay and organic matter to which SB seems to sorb more readily to than AY7. In contrast to anionic exclusion, information on cationic exclusion, as here observed, has not previously been reported in the literature. This can exclude certain contaminant from the matrix domain into the rapid pathways (fractures and macropores) for transport of contamination down to the groundwater.

Modelling of Preferential Flow and Solute Transport in Fractured Vadose Media

Background

For modelling preferential flow and transport in fractured vadose media a variety of approaches have been developed, which are generally based on the commonly accepted mathematical model for uniform transient subsurface water flow in variably saturated porous media the Richards’ equation:

$$\frac{\partial}{\partial x_i}\left(K_{ij} k_{rw} \frac{\partial(\psi + z)}{\partial x_j}\right) \pm Q = \frac{\partial}{\partial t}\left(\theta_s S_w\right) \qquad i, j = 1, 2, 3 \tag{1}$$

and for uniform solute transport of a single species the advection-dispersion equation:

$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial x_i}\left(D_{ij}\frac{\partial c}{\partial x_j} \right) - v_i \frac{\partial c}{\partial x_i} - \Re eact = 0, \quad i, j = 1,2,3 \tag{2}$$

where z is the elevation head [L], $\psi=\psi(x_i,t)$ is the pressure head [L], $K_{ij} = \frac{\rho g}{\mu} k_{ij}$ is the saturated hydraulic conductivity [LT^{-1}], k_{ij} is the permeability of the medium [L^2], ρ is the density of the fluid [ML^{-3}], g is the gravitational acceleration [LT^{-2}], μ is the viscosity of the fluid [$ML^{-1}T^{-1}$], $S_w = \theta / \theta_s$ is the water saturation equal to the water content θ divided by the saturated water content θ [dimensionless], $k_{rw}=k_{rw}(S_w)$ is the relative permeability of the medium with respect to the water saturation S_w [dimensionless], Q is a sink/source term [$L^3L^{-3}T^{-1}$], v_i is the pore water velocity [LT^{-1}], $c=c(x_i,t)$ is the solute concentration, D_{ij} [L^2T^{-1}] is the hydrodynamics dispersion coefficient given by (Bear, 1972) and $\Re eact$ includes all reactive terms (e.g. chemical processes).

The most common approaches can be divided into the following categories: (1) continuum models, (2) discrete fracture network models and (3) hybrid models. The continuum models include single, equivalent, dual porosity, dual permeability, multi porosity and multi permeability models. In a discrete fracture model, fractures are explicitly represented in the model. An intermediate approach is the hybrid models, which combines continuum and the discrete fracture approach.

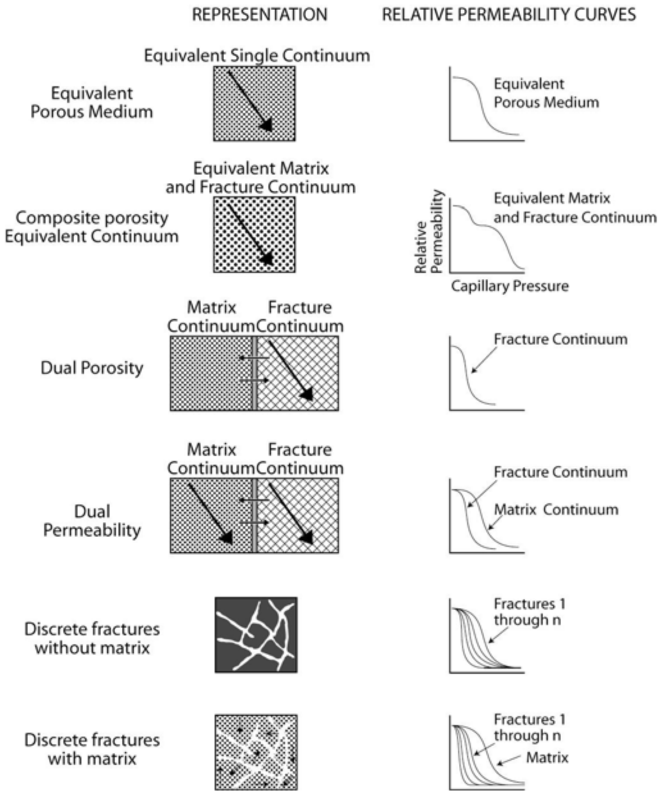

Figure 2. Alternative conceptual models and their relative permeability characteristic curves for flow through fractured rocks (Altman et al., 1996). The arrow in the schematic representation indicates fluid flow through the continuum (matrix and fracture).

The continuum models are based on volume averaging that assumes that processes in fractures and matrix together or individually can be described within a representative elementary volume (REV) of the media. The continuum approach can therefore be applied to fractured media providing that REV can be determined. Two kinds of continuum approaches may be distinguished: (i) phenomenological approaches with which the form of the macroscopic model is postulated on the basis of physical considerations and experimental results; (ii) upscaling methods with which the macroscopic model is rigorously derived by starting with the physical behaviour at the REV's scale (Royer et al., 2002).

The single continuum approach is the simplest approximation to the modelling of flow and transport in fracture media and assumes that an equivalent porous medium can represent either the matrix or the fracture network. This approach may provide valuable insights into the limiting behaviour of a fractured media when e.g. contributions from the matrix can be neglected because of fracture coating (Doughty, 1999).

In the equivalent continuum approach (effective continuum approach, ECM-models), each grid block composites of a fracture and matrix component. ECM uses a composite hydraulic conductivity function assuming that locally, the pressure head in the fracture is equal to the pressure head in the matrix (instantaneous equilibrium). Characteristic curves for the two components of the model are specified independently and only a single effective storativity for both components is assumed resulting in full mixing between the components (Doughty, 1999). This approach is therefore adequate for modelling steady-state flow where strong fracture-matrix interaction is registered. However, the approach makes it impossible to have reliable estimate of the hydraulic head or concentration in a certain point of the domain (Samardzioska, 2005); since it does not provide any insight in the processes of flow and transport in the two different domains – the porous matrix and the fractures. In situations with no detailed knowledge about the fractured media or for large-scale simulations, this simple approach can be a useful tool.

Dual porosity approaches (the mobile-immobile approach) assume that the porous medium consists of two interacting regions, one associated with the inter-aggregate, macropore, or fracture system, and one comprising micropores (or intra-aggregate pores) inside soil aggregates or the rock matrix. Water flow is restricted to the fractures (or inter-aggregate pores and macropores), and in the matrix (intra-aggregate pores or the rock matrix) it is stagnant. Thus, intra-aggregate pores represent immobile pockets that can exchange, retain and store water, but do not permit flow. The dual-porosity formulation for water flow can be based on an extended formulation of the Richards' equation to describe water flow in the fractures and a mass balance equation to describe moisture dynamics in the matrix. Germann and Beven (1985) have suggested an alternative approach for flow in the macropores using a kinematic wave approximation to describe gravitational movement of water in macropores and hereby ignoring capillarity. In general, the dual-porosity models provide a mechanism to account for the delay in the hydraulic response of the matrix caused by fluid that is resident in less permeable matrix blocks. However these models over regularize the geometry of the fracture network, and reliable parameter estimates are difficult to obtain (National Research Council, 1996). The dual porosity models can be used to obtain sufficiently accurate results for practical purposes, especially for modelling domains with large number of fractures with repetitive geometry and similar characteristics, not having to engage into preparation of complicated input data due to the complex geometry of the problem (Samardzioska, 2005). For saturated conditions this approach is widely use for simulating flow and transport in fractured media, but for the vadose zone it is seldom used because it is generally to restrictive (National Research Council, 2001).

The dual-permeability approach is an extension of the dual-porosity approach by allowing for flow through the matrix. Different types of dual-permeability approaches exist (Pruess et al., 1999), the differences being mainly how water flow in and between the two pore regions is descriebed. Approaches to calculating water flow in fractures, macropores or inter-aggregate pores range from those invoking Poiseuille´s equation (Ahuja and Hebson, 1992), the Green and Ampt or Phillip infiltration models (Ahuja and Hebson, 1992; Chen and Wagenet, 1992), the kinemaic wave equation (Germann and Beven, 1985; Jarvis, 1994) and the Richards equation (Gerke and Van Genuchten, 1993a). The model by Gerke and van Genuchten (1993a, 1996) represents a model where Richards equation is applied to each of the two pore regions. This approach requires water retention and hydraulic functions for both pore regions, as well as hydraulic conductivity function of the fracture-matrix interface (Simunek, 2003).

In dual-porosity and dual-permeability models, fracture-matrix interaction is represented by the extent of flow $\Gamma_w = \alpha_w (h_f - h_m)$ between the two pore regions, where h_f and h_m are the pressure heads in the fracture (or inter-aggregate pores and macropores) and matrix (or intra-aggregate pores), respectively and α_w is the controlling mass transfer parameter. Typically, α_w depends on the matrix hydraulic conductivity and average fracture spacing. It is commonly assumed that fracture-matrix flow can occur over the entire fracture surface. A relatively large value for α_w implies little to no fracture flow. To avoid overestimating the degree of matrix-fracture interaction, a reduction factor to decrease the fracture-matrix interface area was introduced by Ho (1997) based on the assumption that if preferential flow occurs in the fracture plan, then only a fraction of the fracture plane is wetted, and the connection area between the fracture and matrix is reduced. To account for fracture coating, Gerke and Van Genuchten (1993b) characterized the fracture-matrix interface by a hydraulic conductivity function, $K_a(\bar{h})$, where $\bar{h}$ is defined as some type of average of pressure heads in the fracture and the matrix (National Research Council, 2001).

Multi porosity and/or multi permeability approach (MINC) are based on the same concept as dual-porosity and dual-permeability approach, but include additional interactive pore regions (Gwo et al., 1995; Hutson and Wagenet, 1995). This allows for greater flexibility, albeit at the expense of requiring more parameters that may also be physically poorly defined. Models like MURF, MURT (Gwo et al., 1995) and TRANSMIT (Hutson and Wagenet, 1995) consider overlapping pore regions and allow for water and solute to exchange between all regions. They can be simplified immediately to dual-porosity/permeability models (Simunek et al., 2003).

Discrete fracture models (also known as the non-homogeneous models) allow for quantification of flow and transport phenomena that are not adequately captured by use of continuum models. A major advantage of the discrete fracture approach is that it can account explicitly (either as a stand-alone flow network or in conjunction with matrix interactions) for the effects of individual fractures on fluid flow and solute transport. In this approach, the cubic law is used to describe flow in the fractures and Richards' equation to describe the flow in the porous matrix (Therrien and Sudicky, 1996). As a consequence, these models have become popular for theoretical studies and even for practical applications, in spite of their computational limitations for large-scale flow and transport (National Research Council, 1996). Practical application of these models can be constrained by the availability of detailed field data; for calibration, such models demand more data than continuum models. Often exact geometry of the fracture network is not usually known, it is difficult to prepare the meshes for complicated geometries and distributions of fractures in the porous media, and for large simulation problems the computational demand is high. Discrete fracture models are suitable for situations where only several fractures or fracture zones are dominating (Samardzioska, 2005).

The hybrid approach (mixed discrete-continuum models) combines the discrete fracture approach with the continuum approach by representing the dominant fractures as 2D-elements embedded in a 3D porous medium that represent the remaining fractures. This setting shares both the simplicity of continuum models while capturing the role of the hydraulic dominate fractures. The numerical model, HydroSphere, represents such an approach (Therrien et al., 2004).

Percolation theory, chaos theory, and the simpler phenomenological models are among the alternative approaches (Pruess et al., 1999) but will not be described further here.

The selection of the appropriate modelling approach for a given application depends on several criteria including the end-results that are required, geometry and scale of the fractured porous media, the field data available and some practical limitations like capabilities of computational resources (Samardzioska et al., 2005).

Contribution

In manuscript III, the preferential flow and transport in a fractured vadose till media is simulated. For the till setting, a discrete fracture set-up in the three-dimensional numerical model HydroSphere was chosen since detailed field-data including exact geometry of the fracture and macropore network exist at a relatively small scale. At first glance the fast flow and transport mechanism registered in the biopores did not conform to assumption of the capillary conceptual model, which holds that flow happens in continuous films, and stops when the water becomes discontinuous. By assuming that this mechanism could be represented by a transient laminar film flow in the macropores and fractures, they were given by discrete 2D elements with properties as close to reality as possible. The outcome of the simulations seemed to predict the complex transient flow and transport pattern in the variably saturated fractured till reasonably well even at high matrix suction. Regrettably, the dynamics of the clayey coating of the biopores at different saturation were not possible to account for in the model. HydroSphere simulated tracer migration into the dead-end biopores in the fall season, which was not observed. HydroSphere has though proven to be a valuable tool in this investigation, even though it does not account fully for the effect of coating and non capillary flow in the biopores and fractures. This transient numerical description of the interaction between the matrix, biopores and tectonic fractures in a variably saturated till is to date not reported.

Characterising Preferential Flow and Transport in Variably Saturated Fractured Media

Background

For a long time preferential flow and transport was not considered, because it was difficult to detect, measure and even more difficult to model. However, in the last 15 to 20 years, efforts have been made to account for this widespread phenomenon and have led to significant understanding of the dynamics of flow and transport processes in these media. Although, predictive capabilities related to real fractured media still remain limited caused by factors like (1) fracture walls are rough; (2) fractures often contain filling material; (3) filling material and the walls themselves are subject to processes of chemical reaction, dissolution, precipitation, mineralization, and/or particle detachment and trapping; (4) leaching in the vadose zone occurs in preferential paths and channels, with a complex interplay between the fractures and the matrix; and (5) flow and transport patterns are often temporally and spatially unstable. Quantitative models of flow and solute transport in the variably saturated fractured media must therefore be predicated on conceptual models of fractured, heterogeneous, and otherwise “disordered” porous media, which account for a variety of preferential flow and transport behaviours.

Parameters for these conceptual models can be obtained, either by direct or indirect measurement, a prior estimation, or some calibration technique (Beven, 1991). Existing experiments rarely provide enough information to fully calibrate the flow and transport models. Hence, experiments or devices must be designed to provide estimates of the many parameters needed in these relatively complex models (Simunek et al., 2003).

Analyses are usually based on measurements of fractures at outcrops, aerial photographs, core samples, and various geophysical techniques, as well as on hydraulic and tracer testing within or between wells. This is due to the very complex nature of fracture networks in the subsurface, and to the virtual impossibility of obtaining detailed structural, hydraulic, and geochemical characterisations of fractures in situ. As a result, studies must often rely on extrapolation of exposed features and indirect measurements, together with subjective considerations, to generate a statistical characterisation of fracture systems (Berkowitz, 2002). The characterisation of the variably saturated fractured media has therefore often been constrained by insufficient spatial resolution of measurements or unknown measuring volume. Direct evidence of preferential flow and transport can though be obtained by staining techniques with dyes or water, which have been extensively used to visualize preferential flow caused by macropore and fracture (Bouma et al., 1977).

Contribution

In manuscript I, II and IV, new ways of characterising the preferential flow and transport in fractured vadose chalk and till are described. In chalk, a combination of detailed fracture mapping and IR-termographic investigations made it possible to outline the hydraulic active fracture-systems. IR-termography has been used for detecting fractures and studying water flow and changes of water content in different media but has until now not been used for studying flow in fractured porous media. In till setting, a combination of detailed characterisation of the different domains (biopores, fractures, matrix and coating) with high spatial resolution concentration maps of two fluorescent tracers made it possible to outline and numerical simulate the tracer migration pattern in detail. The fluorescence imaging procedure described by Aeby et al. (2001) was for the first time used to delineated flow and transport pattern in a fractured till at a depth below 1.5 meter. Based on SEM-images of a tectonic fracture in the till, a pore-size-distribution was estimated and used to estimate characteristic curves for these fractures. Also for the granite, SEM-images were used to describe micromorphology of the fractures and faults and measurement of the fracture aperture (scale~0.01-1 mm). Until now SEM-images has not been used for estimating the above-mentioned characteristics for fractures in till and granite. Additionally, a method for impregnating fractures and faults in the granite with epoxy in situ was developed.

Future perspectives

The following list reflects topics related to my researches that deserve attention to improve our understanding of the preferential flow and transport in fracture variably saturated media.

- The fracture-matrix interaction is a challenging topic for future research. A question like: Does an increase in flux result in an increase in the contact area between matrix and fracture or in the number of fractures actively carrying water, need to be answered. Also the dynamics of different kinds of coatings and its effect on the preferential flow and transport properties need to be investigated.

- Anionic/cationic exclusion of solutes from different geochemical environments of the variably saturated fractured media. Especially the exclusion of contaminant from the low velocity domains (matrix) into the high velocity domains (macropores and fractures) seems to be an important factor in the spreading of contaminants from the soil surface to deeper aquifers.

- Does film flow play a significant role in infiltration? The film flow can be an important mechanism contributing to fast flow in unsaturated fractures and macropores (Tokunaga and Wan, 2001) but based on order-of-magnitude will the rate be insignificant? How to incorporate "fingering", film flow and intermittent (pulse) flow as small scale mechanism into field-scale models remains a difficult challenge.

- In general the development of upscaling procedures for small scale processes to contribution to large-scale phenomena is needed. Continuing field observations, and carefully designed field experiments at different scales, are of critical importance for the development of a sound and defensible understanding of hydrogeology of variably saturated fractured media (Pruess et al., 1999). In this matter, improved sampling techniques must be developed – also in respect to hydraulic active and inactive fractures.

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